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(54) **Polyphenylene ether resin compositions having improved ductile impact strength.**

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(73) Proprietor: **GENERAL ELECTRIC COMPANY**
1 River Road
Schenectady New York 12305 (US)

(72) Inventor: **Holub, Fred Frank**
2263 Preisman Drive
Schenectady New York 12305 (US)
Inventor: **Abolins, Visvaldis**
151 Haddington Lane
Delmar New York 12054 (US)

(74) Representative: **Catherine, Alain et al**
General Electric France Service de Propriété Industrielle 18 Rue Horace Vernet B.P. 76
F-92134 Issy-les-Moulineaux Cedex (FR)

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Description

The polyphenylene ether resins are known to be combinable with alkenyl aromatic polymers to provide thermoplastic compositions which are extrudable and moldable into articles of high heat resistance, good impact strength and hydrolytic stability, and good dimensional stability. The compositions can also be formulated into various types, including flame retardant, reinforced, platable, or foamable grades.

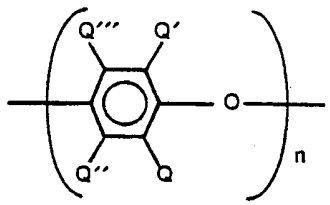
The polyphenylene ether resins and methods of their formation are described in the patent literature, including U.S.—A—3,306,874 and 3,306,875 (Hay), and U.S.—A—3,257,357 and 3,257,358 (Stamatoff). A mixture of polyphenylene ether resin with poly(alkenyl aromatics), including polystyrene, rubber modified polystyrene and styrenic co- and terpolymers, is disclosed in U.S.—A—3,383,435.

Compositions of polyphenylene ether resin and styrene resins have been further modified to improve certain properties by the addition of polyesters. U.S.—A—4,206,154 discloses that the use of a fatty acid-terminated saturated polyester in combination with a polyphenylene ether resin, a styrene resin, a halogenated aromatic flame retardant and ferrocene, results in a composition having enhanced thermal resistance, improved impact strength, and less tendency to undergo dripping when molten. U.S.—A—4,073,765 discloses that the inclusion of an adipic acid based polyester to an admixture of polyphenylene ether resin, styrene resin and titanate filler, enhances the impact strength and tensile elongation.

It has now been discovered that a minor, even a very small amount of (1) a relatively high molecular weight polyfunctional ester of an aliphatic carboxylic acid and a branched polyol, or (2) a specific relatively low molecular weight saturated polyester, or (3) a mixture of the two, when included in flame retardant admixtures and blends of a polyphenylene ether resin, a poly(alkenyl aromatic) and a non-halogenated aromatic phosphate flame retardant agent, sharply increases the ductile impact strength as well as the tensile elongation of articles extruded or molded from the composition. For instance, the articles often possess a ductile impact strength as measured by the Gardner test of greater than 200 in. lbs. (22.59 J), in comparison with less than 200 in. lbs. (22.59 J) for the corresponding composition without the additive(s). Moreover, the heat distortion temperature is maintained, and composition remains flame retardant.

The invention may be practiced with use of a wide variety of materials encompassed within the broader description given above. In the typical case, however, use is made of certain preferred materials which are described below.

The polyphenylene ether resin, which may be designated component (a) of the composition, is normally a homo- or copolymer having units of the formula



wherein Q, Q', Q'' and Q''' are independently selected from the group consisting of hydrogen, halogen, hydrocarbon, halohydrocarbon, hydrocarbonoxy, and halohydrocarbonoxy; and n represents the total number of monomer units and is an integer of at least about 20, and more usually at least 50.

The polyphenylene ether resin can be prepared in accordance with known procedures, such as those described in the above mentioned patents of Hay and Stamatoff, from the reaction of phenols including but not limited to: 2,6-dimethylphenol; 2,6-diethylphenol; 2,6-dibutylphenol; 2,6-dilaurylphenol; 2,6-dipropylphenol; 2,6-diphenylphenol; 2-methyl-6-tolylphenol; 2-methyl-6-methoxyphenol; 2-methyl-6-butylphenol; 2,6-dimethoxyphenol; 2,3,6-trimethylphenol; 2,3,5,6-tetramethylphenol, and 2,6-diethoxyphenol.

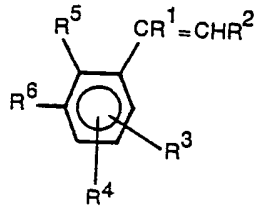
Each of these may be reacted alone to produce the corresponding homopolymer, or in pairs or with still other phenols to produce the corresponding copolymer. Examples of the homopolymer include poly(2,6-dimethyl-1,4-phenylene ether), poly(2,6-diethyl-1,4-phenylene ether), poly(2,6-dibutyl-1,4-phenylene ether), poly(2,6-dilauryl-1,4-phenylene ether), poly(2,6-dipropyl-1,4-phenylene ether), poly(2,6-diphenyl-1,4-phenylene ether), poly(2-methyl-6-tolyl-1,4-phenylene ether), poly(2-methyl-6-methoxy-1,4-phenylene ether), poly(2-methyl-6-butyl-1,4-phenylene ether), poly(2,6-dimethoxy-1,4-phenylene ether), poly(2,3,6-trimethyl-1,4-phenylene ether), poly(2,3,5,6-tetramethyl-1,4-phenylene ether), and poly(2,6-diethoxy-1,4-phenylene ether). Examples of the copolymer include, especially, those of 2,6-dimethylphenol with other phenols, such as poly (2,6-dimethyl-co-2,3,6-trimethyl-1,4-phenylene ether) and poly (2,6-dimethyl-co-2-methyl-6-butyl-1,4-phenylene ether).

For purposes of the present invention, an especially preferred family of polyphenylene ethers include those having alkyl substitution in the two positions ortho to the oxygen ether atom, i.e., those of the above formula wherein Q and Q' are alkyl, most preferably having from 1 to 4 carbon atoms. Illustrative members

of this class are: poly(2,6-dimethyl-1,4-phenylene)ether; poly(2,6-diethyl-1,4-phenylene)ether; poly(2-methyl-6-ethyl-1,4-phenylene)ether; poly(2-methyl-6-propyl-1,4-phenylene) ether; poly(2,6-dipropyl-1,4-phenylene)ether; poly(2-ethyl-6-propyl-1,4-phenylene)ether; and the like.

The most preferred polyphenylene ether resin for purposes of the present invention is poly(2,6-dimethyl-1,4-phenylene)ether.

Component (b) of the composition is an alkenyl aromatic copolymer. The term "alkenyl aromatic polymer" as it is employed in this disclosure is intended to encompass homopolymers, as well as rubber modified high impact varieties, and also copolymers and terpolymers of alkenyl aromatic compounds with one or more other materials. Preferably, the alkenyl aromatic polymer is based at least in part on units of the formula



wherein R^1 and R^2 are selected from the group consisting of lower alkyl or alkenyl groups of from 1 to 6 carbon atoms and hydrogen; R^3 and R^4 are selected from the group consisting of chloro, bromo, hydrogen and lower alkyl of from 1 to 6 carbon atoms; R^5 and R^6 are selected from the group consisting of hydrogen and lower alkyl and alkenyl groups of from 1 to 6 carbon atoms; or R^5 or R^6 may be concatenated together with hydrocarbyl groups to form a naphthyl group.

The above will encompass styrene, as well as homologs and analogs of styrene. Specific examples include, in addition to styrene, chlorostyrene, bromostyrene, alpha-methyl styrene, para-methyl styrene, vinyl styrene, divinylbenzene and vinyl naphthalene. Styrene is especially preferred.

By way of illustration, component (b) can be a polystyrene or other alkenyl aromatic homopolymer which has been modified by admixture or interreaction with a natural or synthetic rubber, for example, polybutadiene, polyisoprene, EPDM rubber or silicone rubber; or it can be a copolymer or terpolymer of styrene or other alkenyl aromatic compound with an elastomeric or other material, such as a block copolymer of styrene and butadiene (for example, AB, ABA, ABAB or ABABA type), including hydrogenated forms of the foregoing, a radial teleblock copolymer of styrene, butadiene and a coupling agent, including hydrogenated forms, terpolymers of acrylonitrile, styrene and butadiene (ABS), styrene-acrylonitrile copolymers (SAN), and a copolymer of styrene and maleic anhydride; or it can also be an alkenyl aromatic copolymer or terpolymer which has been modified with rubber, for example, rubber modified styrene-maleic anhydride copolymer. Many of them are described in the patent literature, including US—A—3 383 435.

The ductile strength improving additive, designated herein also as component (c), is (1) a high molecular weight polyfunctional ester of an aliphatic carboxylic acid and a branched polyol (that is, a di- or polyhydric alcohol), or (2) a low molecular weight saturated polyester, or a mixture of both.

The ester, component (c) (1), is a polyfunctional compound having two or more ester groups, is relatively non-volatile at temperatures of above 100°C., and in general has a molecular weight (weight average) in the range from 300 to 2000.

Suitable polyfunctional esters include those derived from aliphatic carboxylic acid of from 2 to 20 carbon atoms and branched polyols containing from 2 to 4 hydroxyl groups.

Examples of carboxylic acids from which the polyfunctional ester may be prepared are acetic (C_2), propionic (C_3), butyric (C_4), valeric (C_5), caproic (C_6), caprylic (C_8), capric (C_{10}), lauric (C_{12}), myristic (C_{14}), palmitic (C_{16}), stearic (C_{18}) and decosanoic acid (C_{22}).

Examples of branched polyols which may be inter-reacted with the foregoing to form the polyfunctional ester include, especially, pentaerythritol (having four hydroxyl groups), and pinacol (having two hydroxyl groups).

The ester may be prepared in the conventional way such as by heating the carboxylic acid or a derivative such as the acid chloride or anhydride with the polyol in the presence of an amount of a mineral acid, for example, concentrated sulfuric or hydrochloric.

The polyester, component (c) (2), is a polyester based on neopentyldiol and aliphatic dicarboxylic acids having from 2 to 20 carbon atoms and having a weight average molecular weight within the range from 500 to 10,000.

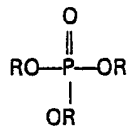
The polyneopentylpolyester is prepared from 2,2-dimethyl-1,3-propanediol (neopentyldiol) and aliphatic dicarboxylic acids having from 2 to 20 carbon atoms.

Examples of dicarboxylic acids are oxalic, malonic, succinic, glutaric, adipic, pimelic, suberic, azelaic and sebacic.

The polyester can be prepared by use of standard techniques known to those skilled in the art. In one procedure, involving direct esterification, the diacid and an excess amount of the diol are heated with stirring in a glass lined or stainless steel reaction vessel, at a temperature of 150 to 250°C., in the presence

of an inert gas, for example, nitrogen. The polyesterification is initially self-catalyzing, due to the presence of the carboxyl groups on the diacid. As the reaction progresses, and as the amount of the diacid is correspondingly reduced, it may be helpful to include a catalyst to maintain the reaction rate. During the reaction, byproduct water and unreacted portions of the diol are distilled off. The progression of the reaction may be monitored by measuring the volume of distillate, or the number of end groups, or the viscosity of samples drawn from the reaction mixture. Care should be taken to avoid extended periods of heating, the use of very high temperatures, or the application of reduced pressure (vacuum), all of which may promote further polymerization to undesirably high molecular weights. Further, details are available in the ENCYCLOPEDIA OF POLYMER SCIENCE AND TECHNOLOGY, John Wiley and Sons, Inc., Volume 11, pages 88—97 (1964).

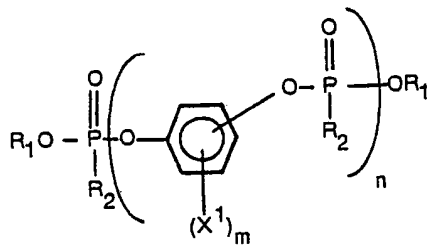
The composition of the invention will also contain as an essential ingredient a flame retardant agent, component (d), which is a non-halogen containing organic phosphate. In the preferred embodiment, the organic phosphate is a compound having the formula

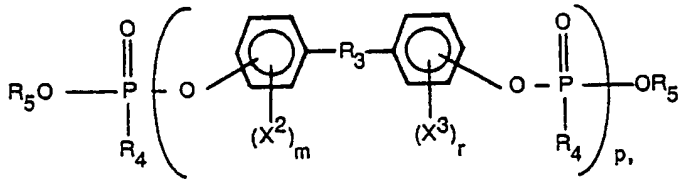


where R is the same or different and is alkyl, cycloalkyl, aryl, alkyl substituted aryl, aryl substituted alkyl, or a combination of any of the foregoing, provided at least one R is aryl.

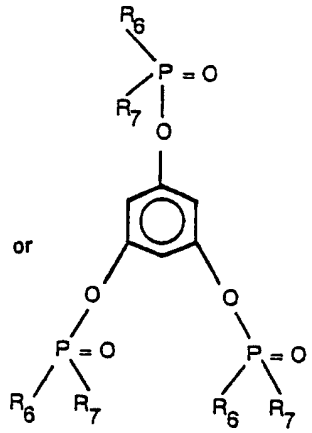
Examples include phenyl bisdodecyl phosphate, phenylbisneopentyl phosphate, phenyl-bis(3,5,5'-trismethylhexyl phosphate), ethyldiphenyl phosphate, 2-ethylhexyldi(p-tolyl) phosphate, bis-(2-ethylhexyl)p-tolylphosphate, tritolyl phosphate, bis-(2-ethylhexyl) phenyl phosphate, tri-(nonylphenyl) phosphate, di(dodecyl)p-tolyl phosphate, tri-cresyl phosphate, triphenyl phosphate, dibutylphenyl phosphate, 2-chloroethyldiphenyl phosphate, p-tolyl bis(2,5,5'-trimethylhexyl) phosphate, 2-ethylhexyldiphenyl) phosphate, and the like. The preferred phosphates are those in which each R is aryl. Especially preferred is triphenyl phosphate, which may be either unsubstituted or substituted with alkyl, for example, isopropylated triphenyl phosphate.

The organic phosphates can also be a difunctional or polyfunctional aromatic compound or polymer of the following formulae:





or



in which R_1, R_3 and R_5 are, independently, hydrocarbon; R_2, R_4, R_6 and R_7 are, independently, hydrocarbon or hydrocarbonoxy; X^1, X^2 and X^3 are halogen; m and r are 0 or integers from 1 to 4, and n and p are from 1 to 30.

Methods of preparation are described in GB—A—2,043,083.

Particular mention is made herein of the bis diphenyl phosphates of resorcinol, hydroquinone and bisphenol-A, respectively, and of their oligomeric and polymeric counterparts.

The polyphenylene ether resin and alkenyl aromatic polymer, components (a) and (b), are admixable in widely variant proportion ranging from, for example, 1:99 to 99:1, but more usually from 5:95 to 95:5, and especially preferably from 20:80 to 80:20, weight ratio. Component (c) need only be added in amounts less than about 50 parts by weight, per 100 parts of (a) and (b) together, to get the described benefits. Typically, only small amounts are employed, and preferably from about 0.1 to about 10 parts by weight for each 100 parts of (a) and (b) combined.

The flame retardant, component (d), is present in amounts conventional for the stated purpose, and generally from about 1 to about 20 parts per 100 parts of (a) and (b) combined.

The components which have been described are further admixable with additional ingredients, if desired, which may be selected from among additives such as stabilizers, for example, metal oxides and sulfides; plasticizers; colorants; mineral fillers, for example, clay, talc, mica, or titanium dioxide; reinforcements, for example, glass flakes, fibers or spheres, or titanate whiskers; melt viscosity adjusters, and so forth. These materials may be added in amounts conventional for the desired purpose and as suited for particular requirements.

After preparation, the compositions can be shaped by extrusion, compression or injection molding, sheeting or other thermal procedures into various articles.

In one procedure, a mixture of the ingredients is extruded at a temperature in the range between 550 (287.8) to 600°F. (315.5°C) and injection molded at 500 (260) to 550°F (287.8°C). The resulting molded articles have good ductility and are especially useful in applications where they must withstand rough handling.

The compositions are suitable for any of the widely divergent uses for which polyphenylene ether resin blends are known. Thus, they are utilizable for automotive exteriors, including grilles, headlamp bezels, wheelcovers and decorative trim; automotive interiors, such as pillar and garnish moldings, center consoles, rear window shelves, and speaker and defogger grilles; automotive instrument panel light clusters, lamp housings, electrical connectors and lamp sockets; major household appliances, such as laundry and dishwasher consoles and lids, motor support housings, drain impellers, ice maker components, compressor covers and air conditioning grilles and fans; small appliances such as coffee makers, irons, food processors, hairsetters, curling irons and shower massages; business machine components, including housings, circuit board brackets, card guides and frames, motor covers, connectors, ductworks and control boxes; electrical construction products, including bus-bar sleeves, electrical cable covers, wiring splice covers, track light insulation and lighting enclosures; and television backs, bases, front/escutcheons and cabinet sides.

The following examples illustrate the invention and benefits which are achieved.

Examples 1—3

Compositions in accordance with the invention were prepared by mixing the ingredients noted below in a blender, extruding the blend through a Werner-Pfleiderer twin screw extruder at a temperature of about 550°F (287.8°C), collecting, cooling and chopping the extrudate into molding pellets, and then injection molding the pellets into test bars using a Newbury injection molding device and an injection temperature of about 500°F (260°C). A control blend was included for purposes of comparison. The compositions and the test results are set forth in the Table below.

TABLE

Ingredients, pts. by wt.	A*	1	2	3
Poly(2,6-dimethyl-1,4-phenylene ether) resin	55	55	55	55
Rubber modified high impact polystyrene	45	45	45	45
Triphenyl phosphate	17	17	17	17
Titanium dioxide	5	5	5	5
Tridecylphosphite(stabilizer)	0.5	0.5	0.5	0.5
Zinc sulfide/zinc oxide	0.15/0.15	0.15/0.15	0.15/0.15	0.15/0.15
Poly(neopentyl adipate)	—	1.0	—	—
Pentaerythritol tetrastearate	—	—	1.0	3.0
Properties				
Tensile strength, psi (MPa)	8,000 (55.16)	8,300 (57.22)	8,200 (56.53)	7,800 (53.78)
Elongation, %	58	76	88	92
Gardner imp. str., in.-lbs. (J)	180 (20.33)	400+ (45.19+)	230 (25.98)	400+ (45.19+)
Heat distortion temp.,-°F./264 psi (°C/1.82 MPa)	192 (88.9)	186 (85.5)	184 (84.4)	180 (82.2)

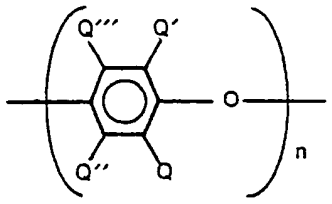
*comparison (control) experiment

As will occur to those skilled in the art, modifications and variations of the invention are possible in light of the above disclosure. For instance, instead of poly(2,6-dimethyl-1,4-phenylene ether) a copolymer such as poly(2,6-dimethyl-co-2,3,6-trimethyl-1,4-phenylene ether) can be employed. Instead of a rubber modified high impact polystyrene, other alkenyl aromatic polymers can be used, such as linear block or radial teleblock copolymers or styrene and butadiene (Shell Chemical's Kraton® and Kraton® G materials or Phillips Petroleum's Solprene® products). Instead of triphenyl phosphate, a substituted derivative such as isopropylated triphenyl phosphate is possible. Mineral, e.g., clay filled or glass reinforced embodiments are also possible. Other non-halogenated flame retardant agents in addition to the organic phosphate can be present to enhance the flame retardancy effect.

Claims

1. A flame retardant thermoplastic composition, comprising
(a) a polyphenylene ether resin;
(b) an alkenyl aromatic polymer;
(c) an additive selected from the group consisting of
(1) a polyfunctional ester of an aliphatic carboxylic acid and a branched polyol having a weight average molecular weight in the range from 300 to 2000.
(2) a polyester based on neopentyldiol and aliphatic dicarboxylic acids having from 2 to 20 carbon atoms and having a weight average molecular weight in the range from 500 to 10000, and
(3) a mixture of (1) and (2); and
(d) a non-halogenated flame retardant agent comprising an organic phosphate.

2. A composition according to Claim 1, in which the polyphenylene ether resin is a homopolymer or copolymer having units of the formula

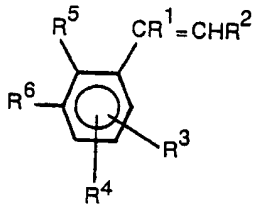


wherein Q, Q', Q'' and Q''' are independently selected from the group consisting of hydrogen, halogen, hydrocarbon, halohydrocarbon, hydrocarbonoxy, and halohydrocarbonoxy; and n represents the total number of monomer units and is an integer of at least about 20.

3. A composition according to Claim 1, in which the polyphenylene ether resin is poly(2,6-dimethyl-1,4-phenylene ether) resin.

4. A composition according to Claim 1, in which the alkenyl aromatic polymer is a homopolymer, a copolymer, a terpolymer, or a rubber modified version of any of the foregoing.

5. A composition according to Claim 1, in which the alkenyl aromatic polymer is derived at least in part from a compound of the formula



wherein R^1 and R^2 are selected from the group consisting of lower alkyl or alkenyl groups of from 1 to 6 carbon atoms and hydrogen; R^3 and R^4 are selected from the group consisting of chloro, bromo, hydrogen and lower alkyl of from 1 to 6 carbon atoms; R^5 and R^6 are selected from the group consisting of hydrogen and lower alkyl and alkenyl groups of from 1 to 6 carbon atoms; or R^5 or R^6 may be concatenated together with hydrocarbyl groups to form a naphthyl group.

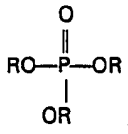
6. A composition according to Claim 1, in which the alkenyl aromatic polymer is a rubber modified high impact polystyrene.

7. A composition according to Claim 1, in which (c) (1) is derived from an aliphatic carboxylic acid of from 2 to about 20 carbon atoms and a branched polyol of from 2 to 4 hydroxyl groups.

8. A composition according to Claim 1, in which (c) (1) is pentaerythritol tetrastearate.

9. A composition according to Claim 1, in which (c) (2) is poly(neopentyl adipate).

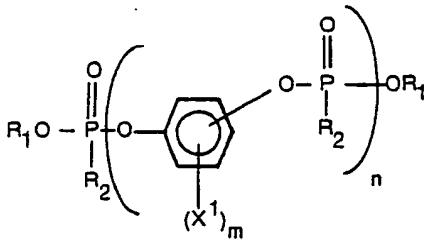
10. A composition according to Claim 1, in which component (d) is a non-halogenated aromatic compound having the formula

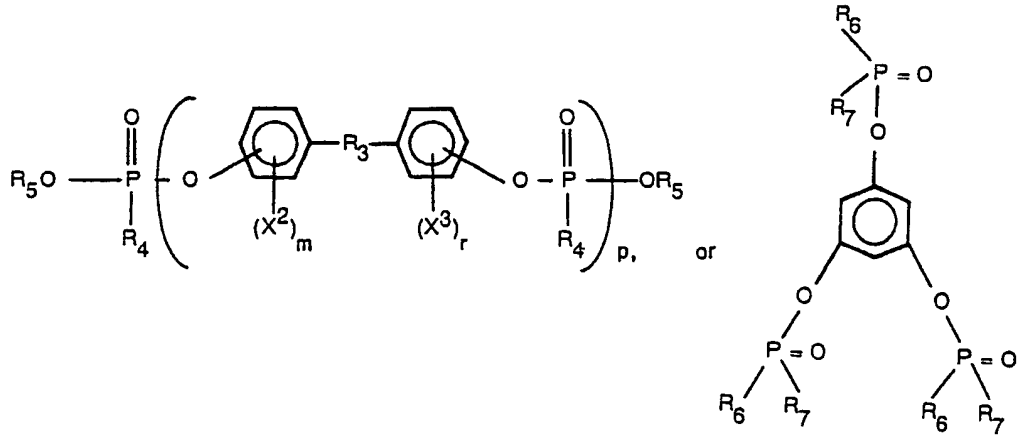


where R is the same or different and is alkyl, cycloalkyl, aryl, alkyl substituted aryl, aryl substituted alkyl, or a combination of any of the foregoing, provided at least one R is aryl.

11. A composition according to Claim 10, in which (d) is triphenyl phosphate.

12. A composition according to Claim 1, in which component (d) is a non-halogenated difunctional or polyfunctional compound or polymer of the formula





in which R_1, R_3 and R_5 are, independently, hydrocarbon; R_2, R_4, R_6 and R_7 are, independently, hydrocarbon or hydrocarbonoxy; X^1, X^2 and X^3 are halogen; m and r are 0 or integers from 1 to 4, and n and p are from 1 to 30.

13. A composition according to Claim 12, in which (d) is a bis diphenyl phosphate of resorcinol, hydroquinone or bisphenol-A.

14. A composition according to Claim 1, in which the weight ratio of (a):(b) is from 5:95 to 95:5.

15. A composition according to Claim 1, in which the weight ratio of (a):(b) is from 20:80 to 80:20.

16. A composition according to Claim 1, which contains from about 0.1 to about 10 parts by weight of (c) for each 100 parts of (a) and (b) combined.

17. A composition according to Claim 1, which contains from about 1 to about 20 parts of (d) per 100 parts of (a) and (b) combined.

18. A composition according to Claim 1, which contains a filler.

19. A composition according to Claim 18, in which the filler is non-reinforcing.

20. A composition according to Claim 19, in which the non-reinforcing filler is titanium dioxide.

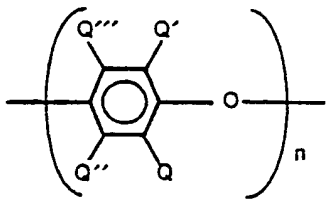
21. A composition according to Claim 1, which contains an effective amount of a stabilizer.

22. A composition according to Claim 21, in which the stabilizer comprises a combination of zinc sulfide and zinc oxide.

Patentansprüche

1. Flammhemmende thermoplastische Zusammensetzung enthaltend
a) ein Polyphenylenätherharz
b) ein alkenylaromatisches Polymer
c) ein Additiv ausgewählt aus der Gruppe bestehend aus
(1) einem polyfunktionellen Ester aus einer aliphatischen Karbonsäure und einem verzweigten Polyol mit einem gewichtsgemittelten Molekulargewicht im Bereich von 300 bis 2000,
(2) einem Polyester auf der Basis von Neopentyldiol und aliphatischen Dikarbonsäuren mit 2 bis 20 Kohlenstoff-Atomen und einem gewichtsgemittelten Molekulargewicht im Bereich von 500 bis 10000 und
(3) einer Mischung aus (1) und (2) und
(d) einem nichthalogenierten, flammhemmenden Mittel, welches ein organisches Phosphat enthält.

2. Zusammensetzung nach Anspruch 1, in welcher das Polyphenylenätherharz ein Homopolymer oder Copolymer mit Einheiten der Formel

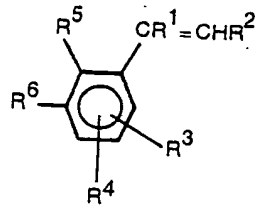


ist, worin Q, Q', Q'', und Q''' unabhängig voneinander ausgewählt sind aus der Gruppe bestehend aus Wasserstoff, Halogen, Kohlenwasserstoff, Halogenkohlenwasserstoff, Kohlenwasserstoffoxy und Halogenkohlenwasserstoffoxy und n die gesamte Zahl der Monomereinheiten dastellt und eine ganze Zahl von wenigstens etwa 20 ist.

3. Zusammensetzung nach Anspruch 1, in welcher das Polyphenylenätherharz Poly (2,6-dimethyl-1,4-phenylenäther) Harz ist.

4. Zusammensetzung nach Anspruch 1, in welcher das Alkenyl aromatische Polymere ein Homopolymer, ein Copolymer, ein Terpolymer oder eine gummimodifizierte Version von einem beliebigen der vorgenannten Stoffe ist.

5. Zusammensetzung nach Anspruch 1, in welcher das alkenylaromatische Polymer wenigstens zum Teil abgeleitet ist von einer Verbindung der Formel



worin R^1 und R^2 ausgewählt sind aus der Gruppe bestehend aus niederen Alkyl oder Alkenyl Gruppen mit 1 bis 6 Kohlenstoffatomen und Wasserstoff; R^3 und R^4 ausgewählt sind aus der Gruppe bestehend aus Chlor, Brom, Wasserstoff und niederem Alkyl mit 1 bis 6 Kohlenstoffatomen; R^5 und R^6 ausgewählt sind aus der Gruppe bestehend aus Wasserstoff niederem Alkyl und Alkenylgruppen mit 1 bis 6 Kohlenstoffatomen; oder R^5 oder R^6 zusammen mit Kohlenwasserstoffgruppen verknüpft sind um eine Naphthylgruppe zu bilden.

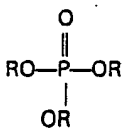
6. Zusammensetzung nach Anspruch 1, in welcher das alkenylaromatische Polymer ein gummimodifiziertes hochschlagfestes Polystyrol ist.

7. Zusammensetzung nach Anspruch 1, in welcher (c) (1) abgeleitet ist von einer aliphatischen Karbonsäure mit 2 bis etwa 20 Kohlenstoffatomen und einem verzweigten Poly mit 2 bis 4 Hydroxylgruppen.

8. Zusammensetzung nach Anspruch 1, in welcher (c) (1) Pentaerythritoltetrastearat ist.

9. Zusammensetzung nach Anspruch 1, in welcher (c) (2) Poly (neopentyladipat) ist.

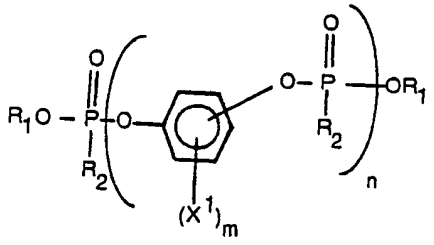
10. Zusammensetzung nach Anspruch 1, in welcher die Komponente (d) eine nichthalogenierte aromatische Verbindung mit der Formel

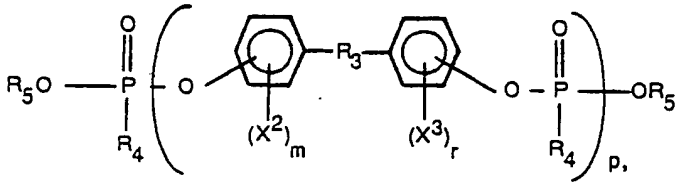


ist, worin die Rest R, gleich oder unterschiedlich sind und Alkyl, Cycloalkyl, Aryl, alkylsubstituiertes Aryl, arylsubstituiertes Alkyl oder eine Kombination aus einer beliebigen der vorgenannten ist mit der Maßgabe, daß wenigstens ein R Rest Aryl darstellt.

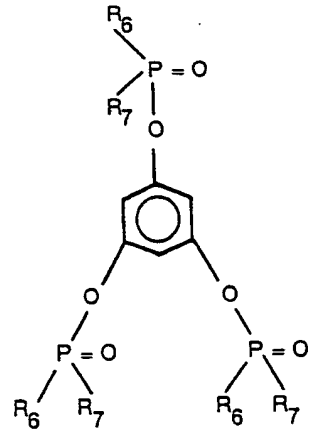
11. Zusammensetzung nach Anspruch 10, in welcher (d) Triphenylphosphat ist.

12. Zusammensetzung nach Anspruch 1, in welcher die Komponente (d) eine nichthalogenierte difunktionelle oder polyfunktionelle Verbindung oder ein Polymer der Formel





oder



ist, worin R_1, R_3 und R_5 unabhängig voneinander Wasserstoff darstellen; R_2, R_4, R_6 und R_7 unabhängig voneinander Kohlenwasserstoff oder Kohlenwasserstoffoxy darstellen; X^1, X^2 und X^3 Halogen sind, m und r 0 oder Ganzzahlen von 1 bis 4 und n und p von 1 bis 30 sind.

13. Zusammensetzung nach Anspruch 12, in welcher (d) ein Bis-Diphenylphosphat von Resorzinol, Hydrochinon oder Bisphenol-A ist.

14. Zusammensetzung nach Anspruch 1, in welcher das Gewichtsverhältnis von (a) zu (b) von 5:95 bis 95:5 beträgt.

15. Zusammensetzung nach Anspruch 1, in welcher das Gewichtsverhältnis von (a) zu (b) von 20:80 bis 80:20 beträgt.

16. Zusammensetzung nach Anspruch 1, welche etwa 0,1 bis 10 Gewichtsteile (c) für jeweils 100 Teile von (a) und (b) zusammen aufweist.

17. Zusammensetzung nach Anspruch 1, welche etwa 1 bis etwa 20 Teile (d) pro 100 Teile von (a) und (b) zusammen aufweist.

18. Zusammensetzung nach Anspruch 1, welche einen Füllstoff enthält.

19. Zusammensetzung nach Anspruch 18, in welcher der Füllstoff nicht verstärkend ist.

20. Zusammensetzung nach Anspruch 19, in welche der nichtverstärkende Füllstoff Titandioxyd ist.

21. Zusammensetzung nach Anspruch 1, in welche eine wirksame Menge eines Stabilisators enthält.

22. Zusammensetzung nach Anspruch 21, in welcher der Stabilisator eine Kombination aus Zinksulfid und Zinkoxyd umfaßt.

Revendications

1. Composition thermoplastique ignifugée comprenant

(a) une résine de poly(éther de phénylène);

(b) un polymère alcénylaromatique;

(c) un additif choisi dans le groupe constitué par

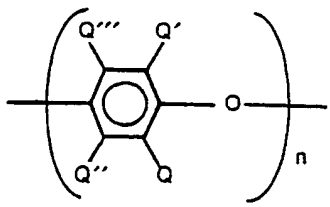
(1) un ester polyfonctionnel d'un acide carboxylique aliphatique et d'un polyol ramifié ayant une moyenne pondérale du poids moléculaire dans la gamme de 300 à 2 000,

(2) un polyester à base de néopentyldiol et d'acides dicarboxyliques aliphatiques ayant de 2 à 20 atomes de carbone et ayant une moyenne pondérale du poids moléculaire dans la gamme de 500 à 10,000, et

(3) un mélange de (1) et de (2); et

(d) un agent ignifuge non halogéné comprenant un phosphate organique.

2. Composition selon la revendication 1, dans laquelle la résine de poly(éther de phénylène) est un homopolymère ou copolymère ayant des motifs de formule:



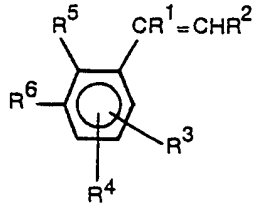
dans laquelle Q, Q', Q'' et Q''' sont indépendamment choisis dans le groupe constitué par un hydrogène, un halogène, un hydrocarbyle, un halogénohydrocarbyle, un hydrocarbonoxy et un

halogénohydrocarbonoxy; et n représente le nombre total des motifs monomères et est un entier d'au moins environ 20.

3. Composition selon la revendication 1, dans laquelle la résine de poly(éther de phénylène) est une résine de poly(2,6-diméthyl-1,4-phénylène).

4. Composition selon la revendication 1, dans laquelle le polymère alcénylaromatique est un homopolymère, un copolymère, un terpolymère ou une version modifiée par un caoutchouc de l'un quelconque de ceux-ci.

5. Composition selon la revendication 1, dans laquelle le polymère alcénylaromatique dérive au moins en partie d'un composé de formule



dans laquelle R^1 et R^2 sont choisis parmi les radicaux alkyles et alcényles inférieurs de 1 à 6 atomes de carbone et l'hydrogène; R^3 et R^4 sont choisis parmi chloro, bromo, l'hydrogène et un alkyle inférieur de 1 à 6 atomes de carbone; R^5 et R^6 sont choisis parmi l'hydrogène et les radicaux alkyles et alcényles inférieurs de 1 à 6 atomes de carbone; ou R^5 ou R^6 peut être lié avec des radicaux hydrocarbyles pour former un radical naphtyle.

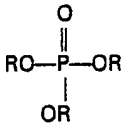
6. Composition selon la revendication 1, dans laquelle le polymère alcénylaromatique est un polystyrène choc modifié avec du caoutchouc.

7. Composition selon la revendication 1, dans laquelle (c) (1) dérive d'un acide carboxylique aliphatique de 2 à environ 20 atomes de carbone et d'un polyol ramifié ayant 2 à 4 radicaux hydroxyles.

8. Composition selon la revendication 1, dans laquelle (c) (1) est le tétrastéarate de pentaérythritol.

9. Composition selon la revendication 1, dans laquelle (c) (2) est un poly(adipate de néopentyle).

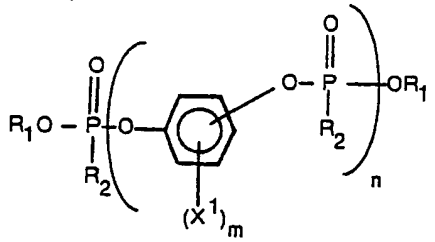
10. Composition selon la revendication 1, dans laquelle le composant (d) est un composé aromatique non halogéné répondant à la formule



dans laquelle les symboles R sont semblables ou différents et représentent un alkyle, un cycloalkyle, un aryle, un aryle à substitution alkyle, un alkyle à substitution aryle ou une combinaison quelconque de ceux-cis, sous réserve qu'au moins un R soit un aryle.

11. Composition selon la revendication 10, dans laquelle (d) est le phosphate de triphényle.

12. Composition selon la revendication 1, dans laquelle le composant (d) est un composé polymère difonctionnel ou polyfonctionnel non halogéné de formule



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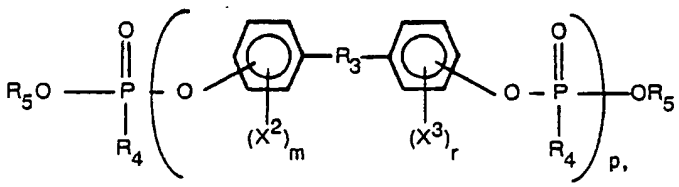

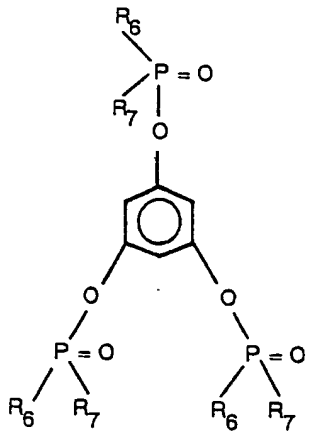

dans laquelle R^1, R^3 et R^5 sont indépendamment un hydrocarbyle; R^2, R^4, R^6 et R^7 sont indépendamment un hydrocarbyle ou un hydrocarbonoxy; X^1, X^2 et X^3 sont un halogène; m et r sont 0 ou des entiers de 1 à 4 et n et p ont une valeur de 1 à 30.

13. Composition selon la revendication 12, dans lauqelle (d) est un phosphate de bis diphényle et de résorcinol, d'hydroquinone ou de bisphénol-A.

14. Composition selon la revendication 1, dans laquelle le rapport pondéral (a)/(b) est de 5/95 à 95/5.

15. Composition selon la revendication 1, dans laquelle le rapport pondéral (a)/(b) est de 20/80 à 80/20.

16. Composition selon la revendication 1, qui contient d'environ 0,1 à environ 10 parties en poids de (c) pour 100 parties de (a) et (b).

17. Composition selon la revendication 1, qui contient d'environ 1 à environ 20 parties en poids de (d) pour 100 parties de (a) et (b).

18. Composition selon la revendication 1, qui contient une charge.

19. Composition selon la revendication 18, dans laquelle la charge n'a pas d'effet de renforcement.

20. Composition selon la revendication 19, dans laquelle la charge n'ayant pas d'effet de renforcement est du dioxyde de titane.

21. Composition selon la revendication 1, qui contient une quantité efficace d'un stabilisant.

22. Composition selon la revendication 21, dans laquelle le stabilisant comprend une combinaison de sulfure de zinc et d'oxyde de zinc.